Continuous soil gas monitoring related to CCS – Lessons learned from a 5-year case study

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The development of adequate monitoring strategies for CO_2 storage sites is one of the most vigorously discussed subjects related to carbon capture and storage (CCS), both in the public and in the scientific community. Public acceptance of CO_2 sequestration will only be achieved if secure and comprehensible monitoring methods for the natural environment are applied in a transparent way. Deep geological monitoring is mostly related to large scale migration of injected carbon dioxide in the storage formations. In contrast, detection and quantification of different gas species in the vadose zone of soil column and/or in the atmospheric boundary layer is a key method in many fields of near-surface environmental research and geohazards as well as CCS. As CO_2 soil gas concentrations vary over broad ranges, reliable statements on CO_2 seepage can only be made by using continuous long-term gas concentration measurements. We describe lessons learned from the first continuous monitoring program applied on a proposed CO_2 storage site in the Altmark area (Germany).

We will focus on our technical experiences, data interpretation and recommendations for further monitoring programs related to CCS. The most important topic is the reliability of a single station's data quality. Each selected site needs a thorough pre-investigation, e.g. regarding the depth of the biologically active zone and potential free water level. Based on our long lasting experience from field studies and including recent monitoring programs, we strongly recommend that baseline monitoring schemes for CO_2 storage sites should start at least 3 years before the gas injection into the reservoir. We will show that vadose zone monitoring can be considerably more sensitive to small seepage rates than surface flux measurements under favourable conditions but will also address pitfalls and limitations of the method.

Sorption of uranium and neptunium onto diorite from Äspö HRL

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Granitic subsurface environments are considered as potential host rock formations for the deep underground disposal of radioactive waste. The retention behavior of the crystalline rock diorite from the Äspö Hard Rock Laboratory (HRL, Sweden) towards the redox-sensitive actinides U and Np was studied by means of batch sorption experiments. The influence of various parameters, such as grain size (0.063 – 0.2 mm, 0.5 – 1 mm, 1 – 2 mm), temperature (25 and 10°C), atmosphere and sorption time (5 to 108 days) was studied using a synthetic Äspö groundwater (pH 7.8, $I = 0.178$ M) as background electrolyte. For U(VI), sorption isotherms were recorded (5×10^{-9} M to 7×10^{-5} M). Distribution coefficients, K_d values, were determined. The K_d values decrease with increasing grain size of the diorite and with decreasing temperature. The K_d values determined under oxic conditions are lower than those determined under anoxic conditions.

In the U sorption system, the speciation of U(VI) in solution and thus, its sorption onto diorite is strongly influenced by the groundwater composition. It was found by time-resolved laser-induced fluorescence spectroscopy that $Ca_2UO_2(CO_3)_3(aq)$ is the dominating species in solution. Hence, calcium and carbonate ions leached out of diorite significantly affect U(VI) sorption. Kinetic experiments showed that sorption equilibrium ($K_d = 1.44 \pm 0.30$, 1 – 2 mm fraction) is reached relatively fast (after 10 to 20 days). As predominant surface species on diorite, $UO_2(CO_3)_3^{4-}$ was identified by in situ time-resolved attenuated total reflection Fourier-transform infrared (ATR FT-IR) spectroscopy. About 50% of the sorbed U can be desorbed with Äspö ground water. This part occurs predominantly as U(VI) (94%) as shown by TTA solvent extraction.

In the Np sorption system, the effect of the groundwater composition on speciation and sorption behavior is weak. By ATR FT-IR spectroscopy, NpO_2^+ was detected in solution. During sorption experiments under anoxic conditions (up to 108 days), Np(V) is reduced to Np(IV) by the Fe(II) of the diorite which leads to a very strong Np sorption (95% sorption for $[Np]_0 = 1 \times 10^{-6}$ M, S/L = 200 g/L). Consequently, only 5-6% of the sorbed Np can be desorbed with Äspö ground water. This small amount was identified as Np(V) by TTA solvent extraction.